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University of Rochester
Office of Research & Project Administration
518 Hylan Bldg., River Campus Box 270140
Rochester, NY 14627 -

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13. ABSTRACT (Maximum 200 words)

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FINAL PROGRESS REPORT "Polarized single-photons on demand - a new source for quantum information

ABSTRACT

The goal of this project is the development of an efficient source, operating at room temperature, of deterministically polarized single photons on demand for quantum information. Our main results are as follows: (1) first demonstration of emitter fluorescence antibunching in liquid crystal hosts; (2) first demonstration of a definite polarization in fluorescence from single emitters (dye molecules) at room temperature both in oligomeric and monomeric planar-aligned nematic liquid crystals; (3) avoiding dye bleaching during more than one hour of cw, 532-nm excitation by special host treatment; (4) first single-dye-molecule/single semiconductor colloidal quantum dot fluorescence imaging in 1-D photonic bandgap chiral nematic liquid crystals; (5) current single-photon source on demand efficiency using pulsed laser excitation of 1-D photonic bansgap doped liquid crystal structure is ~ 10%. We have published 18 journal papers, submitted one patent application, delivered 30 conference and meeting presentations with six invited presentations among them.

List of papers submitted or published that acknowledge ARO support during this reporting period. List the papers, including journal references, in the following categories:

(a) Papers published in peer-reviewed journals (N/A for none)

1. “Room temperature source of single photons of definite polarization”, S. G. Lukishova, A. W. Schmid, R. Knox, P. Freivald, L. Bissell, R.W. Boyd, C.R. Stroud, Jr, K. L. Marshall, J. Modern Optics, Special issue on Single Photon: Detectors, Applications and Measurements Methods, 13 pages, will be published in 2006.

2. “Near-field optical microscopy of defects in cholesteric oligomeric liquid crystal films”, S.G. Lukishova. and A.W. Schmid, Molec. Cryst. Liq. Cryst., 454, 417-423 (2006).

3. “Single-photon source for quantum information based on single dye molecule fluorescence in liquid crystal host”, S.G. Lukishova, A.W. Schmid, R.P. Knox, P. Freivald, A. McNamara, R.W. Boyd, C.R. Stroud, Jr., K.L. Marshall, Mol. Cryst. Liq. Cryst., 454, 403-416 (2006).

4. “Far-field patterns from dye-doped planar-aligned nematic liquid crystals under nanosecond laser irradiation”, S.G. Lukishova, N. Lepeshkin, R.W. Boyd, K. Marshall, Molec. Cryst. Liq. Cryst., 453, 393-401 (2006).

5. “Phase-difference equations: a calculus for quantum revivals”, D. L. Aronstein and C. R. Stroud, Jr., Laser Physics, 15, 1496 (2005).

6. “Deterministic secure communications using two-mode squeezed states”, A.M. Marino and C.R. Stroud, Jr., Phys. Rev. A 74, 022315 (2006).

7. “Robust multipartite multilevel quantum protocols”, H. Nihira and C. R. Stroud, Jr. , Phys. Rev. A 72, 022337 (2005).

8. “Transfer of amplitude and phase modulation to a different wavelength using coherently prepared sodium vapor”, R. S. Bennink, A. M. Marino, V. Wong, R. W. Boyd, and C. R. Stroud, Jr. , Phys. Rev. A 72, 023827 (2005).

9. “Dye-doped cholesteric-liquid-crystal room-temperature single photon source”, S.G. Lukishova, A.W. Schmid, Ch. M. Supranowitz, N. Lippa, A. J. McNamara, R.W. Boyd, and C.R. Stroud, J. of Modern Optics, Special Issue on Single Photon: Detectors, Applications and Measurements Methods, 51, No 9-10, 1535-1547 (2004).

10. “Pas de Deux for Atomic Electrons”, C. R. Stroud, Jr., Science 303, 778 (2004).

11. “Pixel Entanglement: Experimental Realization of Optically Entangled d=3 and d=6 Qudits”, M.N. O'Sullivan-Hale, I. Ali Khan, R.W. Boyd, J.C. Howell, Phys. Rev. Lett. 94, 220501 (2005).

12. “Realization of the Einstein-Podolsky-Rosen Paradox Using Momentum-and Position-Entangled Photons from Spontaneous Parametric Down Conversion”, J.C. Howell, R.S. Bennink, S.J. Bentley, and R.W. Boyd, Phys. Rev. Lett. 92, 210403 (2004).

13. “Quantum and Classical Coincidence Imaging”, R.S. Bennink, S.J. Bentley, R.W. Boyd, and J.C. Howell, Phys. Rev. Lett., 92, 033601 (2004).

14. “Image formation using quantum-entangled photons”, R.W. Boyd, R.S. Bennink, S.J. Bentley, J.C. Howell, published in the “Optics in 2004” section of Optics and Photonics News, December, 2004.

15. “Room-temperature single photon source: single dye molecule fluorescence in liquid crystal host”, S.G. Lukishova, A.W. Schmid, A.J. McNamara, R.W. Boyd, and C.R. Stroud, IEEE J. of Selected Topics in Quantum Electronics, Special issue on Quantum Internet Technologies, 9, N 6, 1512-1518 (2003).

16. “Cumulative birefringence effects of nanosecond laser pulses in dye-doped planar nematic liquid crystal layers”, S.G. Lukishova, R.W. Boyd, N. Lepeshkin, and K.L. Marshall, J. Nonl. Opt. Phys. & Mater., Special issue on Novel Optical Materials and Applications, 11, December, 341-350 (2002).

Number of Papers published in peer-reviewed journals: 16.00

(b) Papers published in non-peer-reviewed journals or in conference proceedings (N/A for none)

1. “Deterministically polarized fluorescence from uniaxially aligned single dye molecules”, S. G. Lukishova, A. W. Schmid, R. Knox, P. Freivald, L. Bissell, R.W. Boyd, C.R. Stroud, Jr, K. L. Marshall, LLE Review, Quarterly Report, University of Rochester, Laboratory for Laser Energetics, January-March , DOE/SF/19460-667, 106, 102-107 (2006).

2. “Demonstration of a room-temperature single-photon source for quantum information: single-dye-molecule fluorescence in a cholesteric liquid crystal host”, S.G. Lukishova, A.W. Schmid, A. J. McNamara, R.W. Boyd, and C.R. Stroud, LLE Review, Quarterly Report, DOE/SF/19460-485, Laboratory for Laser Energetics, University of Rochester, 94, Jan-March, 97-106 (2003).

Number of Papers published in non peer-reviewed journals: 2.00

(c) Presentations

1. (Invited): Dye-doped liquid-crystal room-temperature single photon source, S.G. Lukishova, A.W. Schmid, Ch. M. Supranowitz, A.J. McNamara, R.W. Boyd, C.r. Stroud, Jr., Laser Science XX/Frontiers in Optics 2004, paper LMF2, October 10-14, Rochester, NY.

2.(Invited): "Quantum Optics and Quantum Information Teaching Laboratory Course", S.G. Lukishova, C.R. Stroud, Jr., L. Bissell, A. K. Jha, L. Elgin, Frontiers in Optics, Special Symposium "Quantum Optics and Quantum Information Teaching Experiments", Rochester, NY, October 12 (2006).

3."Deterministically Polarized, Room Temperature Source of Single Photons", S.G. Lukishova, Workshop on Linear Optical Quantum Information Processing, Baton Rouge, Louisiana, 10-12 April 2006.

4. "Quantum Optics and Quantum Information Teaching Experiments", S.G. Lukishova, Workshop on Linear Optical Quantum Information Processing, Baton Rouge, Louisiana, 10-12 April 2006.

5. (Invited): "Quantum optical engineering, C. R. Stroud, Jr., Institute of Optics 75th Anniversary Symposium, Optical Society of America, Rochester, NY, October, 2004.

6. "An efficient, room temperature single-photon source", S. G. Lukishova, A. W. Schmid, R. Knox, P. Freivald, L. Bissell, R.W. Boyd, C.R. Stroud, Jr, K. L. Marshall, Industrial Associates Meeting, April 2006, Rochester NY.

7. "Room-temperature single-photon source for quantum information based on single-dye molecules in liquid crystal hosts", S.G. Lukishova, MRS Rochester Chapter Meeting, April 2006, Rochester NY.

8. (Invited): "Quantum weirdness", C.R. Stroud, Jr., Lecture, Marshall University, October 13, 2005.

9. "Room-temperature single-photon source: dye fluorescence in a liquid crystal host", S.G. Lukishova, A.W. Schmid, C.M. Supranowitz, A.J. McNamara, R.W. Boyd, C.R. Stroud, MURI Review Meeting, July 2003, Rochester NY, Poster.

10. "Quantum information teaching laboratory", A.K. Jha, L. Elgin, S.G. Lukishova, C.R. Stroud, MURI Review Meeting, July 2003, Rochester NY, poster.

11. "Quantum optics/quantum information teaching laboratory", A.K. Jha, L. Elgin, S.G. Lukishova, C.R. Stroud, Industrial Associates Meeting, November 2003, Rochester NY, poster.

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Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

(Invited): "Feedback-free, single-beam pattern formation in dye-doped liquid crystals", N. Lepeshkin, S.G. Lukishova, R.W. Boyd, K.L. Marshall, SPIE Optics & Photonics, August 2006, San Diego, CA.

Number of Non Peer-Reviewed Conference Proceeding publications (other than abstracts): 1

Peer-Reviewed Conference Proceeding publications (other than abstracts):

1. “Deterministically polarized, room temperature source of single photons based on single-emitter fluorescence in aligned liquid crystal hosts”, S.G. Lukishova, A.W. Schmid, R.P. Knox, P. Freivald, L. Bissell. R.W. Boyd, C.R. Stroud, Jr, K.L. Marshall, Technical Digest, CLEO/QELS Conference, May 21-26, 2006, Long Beach, CA, paper JWB97, 2 pages, 2006.

2. “Deterministically polarized, room temperature source of single photons”, S.G. Lukishova, A.W. Schmid, R. Knox, P. Freivald, S. Schrauth, L. Bissell, R.W. Boyd, C.R. Stroud, Jr, K.L. Marshall, Second Single-Photon Workshop (SPW2005): Sources, Detectors, Applications and Measurement Methods, October 24-26, 2005, Teddington, UK, pp. 15-16 (2005).

3. “Quantum optics and quantum information teaching laboratory at the Institute of Optics, University of Rochester”, S.G. Lukishova, C.R. Stroud, Jr., A.K. Jha, L. Elgin, S. Schrauth, L. Bissell, Second Single-Photon Workshop (SPW2005): Sources, Detectors, Applications and Measurement Methods, October 24-26, 2005, Teddington, UK, pp. 83-84 (2005).

4. “Oligofluorene as a new high-performance dye for cholesteric liquid crystal lasers”, K. Dolgaleva, S. K. Wei, A. Trajkovska, S. Lukishova, R.W. Boyd, S.- H. Chen, Technical Digest, Frontiers in Optics/Laser Science 2006, October 8-12, 2006, Rochester, NY, paper FThD4.

5. “Cholesteric liquid crystal laser using an oligofluorene for high performance and spectral purity”, K.-H. Wei, K. Dolgaleva, A. Trajkovska, S. Lukishova, R.W. Boyd, S.H. Chen, Organic Photonics and Electronics, October 9-11, 2006, Rochester, NY, paper OPTuD16.

6. Single-photon source for quantum information based on single dye molecule fluorescence in liquid crystal host, S.G. Lukishova, A.W. Schmid, R.P. Knox, P. Freivald, C.M. Supranowitz, A.J. McNamara, R.W. Boyd, C.R. Stroud, Jr., K.L. Marshall, Book of Abstracts of the 11th International Topical Meeting on Optics of Liquid Crystals (2-7 October 2005, Sand Key, Florida), p. 26 (2005).

7. Feedback-free hexagon pattern formation with liquid crystals and isotropic liquids, S.G. Lukishova , N. Lepeshkin, R.W. Boyd, K.L. Marshall, Book of Abstracts of the 11th International Topical Meeting on Optics of Liquid Crystals (2-7 October 2005, Sand Key, Florida), pp. 42-43 (2005).

8. Near-field optical microscopy of cholesteric oligomeric liquid crystal layers, S.G. Lukishova and A.W. Schmid, Book of Abstracts of the 11th International Topical Meeting on Optics of Liquid Crystals (2-7 October 2005, Sand Key, Florida), pp. 112-113 (2005).

9. Deterministically polarized fluorescence from single dye molecules aligned in liquid crystal host, S.G. Lukishova, A.W. Schmid, R.P. Knox, P. Freivald, R.W. Boyd, C.R. Stroud, Jr, K.L. Marshall, Technical Digest, paperJWH1-2, IQEC 2005 (11-15 July 2005, Tokyo, Japan).

10. Deterministically polarized fluorescence from single dye molecules aligned in liquid crystal host, S.G. Lukishova, A.W. Schmid, R.P. Knox, P. Freivald, R.W. Boyd, C.R. Stroud, Jr, K.L. Marshall, Technical Digest, paper QTuE6, QELS 2005, May 2005. Baltimore, MD).

11. Single-beam light-induced phenomena in dye-doped liquids and liquid crystals, S. Lukishova, Technical Digest, QWAB3-P54, IQEC 2005 (11-15 July 2005, Tokyo, Japan).

12. “Quantum optics and quantum information teaching laboratory at the Institute of Optics, University of Rochester”, S.G. Lukishova, C.R. Stroud. Jr., A.K. Jha, L. Elgin, S. Schrauth, Technical Digest, Frontiers in Optics/Laser Science 2005, Forum on Education, October 16-20, 2005, Tucson, Arizona, paper FThL2 (2 pages).

13. Deterministically polarized single-photon source, S.G. Lukishova, A.W. Schmid, R. Knox, P. Freivald, R.W. Boyd, C.R. Stroud, Jr, Abstract Digest, Quantum Optics II, Cozumel, Mexico, December 6-9, 2004. For presentation see website http://speckle.inaoep.mx/QOII/ppts/Lukishova.pdf.

14. Deterministically polarized, room-temperature single-photon source: single dye molecule fluorescence in liquid crystal host, S. G. Lukishova, A. W. Schmid, C. M. Supranowitz, N. Lippa, A. J. McNamara, R. W. Boyd, C. R. Stroud, IQEC04, San Francisco, CA (May 16-21, 2004), Technical Digest CD-ROM, paper IThG5, 2004.

15. (Invited): Quantization via fractional revivals, C. R. Stroud, Jr., Abstract Digest, Quantum Optics II, Cozumel, Mexico, December 6-9, 2004.

16. Room temperature single-photon source: laser control of single dye molecule fluorescence in photonic-band-gap liquid crystal host, S.G. Lukishova, A.W. Schmid, A. J. McNamara, R.W. Boyd, and C.R. Stroud, Quantum Electronics and Laser Science Conference QELS 2003

(Baltimore, June 2003), Technical Digest, paper JWC7.

17. Efficient room temperature single-photon source for quantum information: single dye molecule fluorescence in photonic-band-gap cholesteric liquid crystal host, S.G. Lukishova, A.W. Schmid, A. J. McNamara, R.W. Boyd, and C.R. Stroud, Frontiers in Optics, the 87th OSA Annual meeting, Technical Digest, paper WF6, Oct. 5-9, 2003, Tucson, Arizona.

18. “Dye-doped cholesteric liquid crystal single photon source”, S.G. Lukishova, A.W. Schmid, A.J. McNamara, R.W. Boyd, and C.R. Stroud, NIST Workshop on Single Photons: Detectors, Applications and Measurements Methods (Gaithersburg, MD, March 31 – April 1 2003).

Number of Peer-Reviewed Conference Proceeding publications (other than abstracts): 18

(d) Manuscripts

1. “Feedback-free, single-beam pattern formation in dye-doped liquid crystals”, N. Lepeshkin, S.G. Lukishova, R.W. Boyd, K.L. Marshall, Proceed. SPIE, Optics & Photonics, August 2006, San Diego, CA, accepted.

2. “Vibrant Imaging: 15-fold dynamic-range enhancement by odd-halves AFM-cantilever overtones”, A.W. Schmid, S.G. Lukishova, (in submission).

Number of Manuscripts: 2.00

Number of Inventions:

Graduate Students

NAME	PERCENT_SUPPORTED	
Luke Bissell (supported by SMAR	0.00	No
FTE Equivalent:	**0.00**	
Total Number:	**1**	

Names of Post Doctorates

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Names of Faculty Supported

NAME	PERCENT_SUPPORTED	National Academy Member
Svetlana G. Lukishova	0.42	No
Robert W. Boyd	0.03	No
Carlos R. Stroud	0.03	No
FTE Equivalent:	**0.48**	
Total Number:	**3**	

Names of Under Graduate students supported

NAME	PERCENT_SUPPORTED	
Sean White (supported by NSF RE		No
Andrew McNamara - 0.24% (0.0(		No
Russell Knox - 0.24% (0.0024)		No
Christopher Supranowitz - 0.24% (		No
Samuel Schrauth (student researc		No
FTE Equivalent:		
Total Number:	**5**	

Names of Personnel receiving masters degrees

NAME

Total Number:

Names of personnel receiving PHDs

NAME

Total Number:

Names of other research staff

NAME	PERCENT_SUPPORTED
Nadine Lippa - high-school intern (	No
Patrick Frievald - research associa	No
FTE Equivalent:	
Total Number:	**2**

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5 **Efficient room-temperature source of polarized single photons**

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5a: Svetlana G. Lukishova, Robert W. Boyd, Carlos R. Stro

5f-1a: University of Rochester

5f-c: the Institute of Optics, University of Rochester

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Sincerely,

Dr. Svetlana Lukishova, PI
Senior Scientist
The Institute of Optics
University of Rochester
Phone: 585/275-8007
E-mail: sluk@lle.rochester.edu

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13. ABSTRACT (Maximum 200 words) The goal of this project is the development of an efficient source, operating at room temperature, of deterministically polarized single photons on demand for quantum information. Our main results are as follows: (1) first demonstration of emitter fluorescence antibunching in liquid crystal hosts; (2) first demonstration of *a definite* polarization in fluorescence from *single* emitters (dye molecules) at room temperature both in oligomeric and monomeric planar-aligned nematic liquid crystals; (3) avoiding dye bleaching during more than one hour of cw, 532-nm excitation by special host treatment; (4) first single-dye-molecule/single semiconductor colloidal quantum dot fluorescence imaging in 1-D photonic bandgap chiral nematic liquid crystals; (5) current single-photon source on demand efficiency using pulsed laser excitation of 1-D photonic bansgap doped liquid crystal structure is ~ 10%. We have published 18 journal papers, submitted one patent application, delivered 30 conference and meeting presentations with six invited presentations among them.

14. SUBJECT TERMS
Single-photon source on demand, single dye molecules, semiconductor colloidal quantum dots, fluorescence antibunching, nematic liquid crystals, 1-D photonic band gap, chiral nematic liquid crystals

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Polarized single-photons on demand - a new source for quantum information

Dr. Svetlana G. Lukishova, Principal Investigator
Dr. Robert W. Boyd and Dr. Carlos R. Stroud – co-Principal Investigators

1 August 2002 – July 31 2006
DAAD 19-02-1-0285

Final Progress Report

Table of Content

1. Statement of the problem studied

The goal of this project is the development of an efficient source, operating at *room temperature*, of *deterministically polarized* single photons on demand. The fields of quantum communication and quantum computing need a simple reliable source of single photons propagating in a given direction with a definite polarization in order to function efficiently. It is important that within a specified time interval, there be one and only one photon in the given direction and polarization. Weak, attenuated sources are not satisfactory because, in order to attenuate the source sufficiently so that two simultaneous photons are very unlikely, the probability of no photons at all becomes large. Similarly, if the single photon is radiated into a large solid angle it is quite difficult to collect it into the optical system with good efficiency. If the photon has unknown polarization, then filtering it through a polarizer to produce the desired polarization will cause a loss of a large percentage of the photons so that again we have zero photons much of the time. It is a difficult engineering problem to produce the ideal source.

As the result of work on this project, we demonstrated an important step along the way to developing such a source. We placed isolated emitters (dye molecules and/or colloidal semiconductor quantum dots) into a liquid crystal matrix that has several desirable properties as an environment for the molecules. The molecules themselves have a very large absorption cross section for laser radiation and then, once excited, they emit a photon Stoke-shifted in frequency by tens of nanometers from the exciting pulse. The lifetime for the photon emission is a few nanoseconds. By exciting the molecule with a sub-nanosecond pulse of sufficient energy, we can guarantee that, with good probability, the single dye molecule will emit a single photon in a few-nanosecond time window after the excitation.

The liquid crystal hosts which can exist both as monomers (fluid media) and oligomers/ polymers serve two purposes. Nematic liquid crystals align the dye molecules along a definite axis. Such aligned molecules would normally emit in a dipole pattern which is a big improvement over the more common situation in which the molecular axes, and thus the dipole patterns, are randomly oriented in space. The chiral nematic (cholesteric) liquid crystals further improve the situation by providing a photonic crystal environment into which the molecules will emit. This environment is provided by supplying a torque to the medium as it is being laid down. The liquid crystals are dichroic, and this torque causes the principal axis of the index of refraction to rotate periodically about an axis perpendicular to the thin layer sample. By making the pitch angle of the rotation of the order of the wavelength of the emitted photon, we can form a photonic crystal that will modify the radiation pattern of the molecule. The photon will be preferentially emitted along the rotation axis. These 1-D photonic band-gap structures in cholesteric liquid crystals possess three main advantages over conventional 1-D photonic crystals:

(1) Because the refractive index *n* varies gradually rather than abruptly in cholesterics, there are no losses into the waveguide modes, which in the case of conventional 1-D photonic crystals, arise from total internal reflection at the boundary between two consecutive layers with a different *n*. These waveguide losses can reach ~20%.

(2) High polarization purity for circular polarization of definite handedness can be reached for single emitters with both polarized and unpolarized emission;

(3) Liquid crystal hosts can provide tunability of a single-photon source by external fields.

In addition to emitter alignment and self-assembled structures with photonic band-gap properties, such a host *with special treatment* (oxygen depletion) can protect the emitters from bleaching.

2. Summary of the most important results

We have achieved significant results under the current award. We have published 18 journal papers, submitted one patent application, delivered 30 conference and meeting presentations with six invited presentations among them (see Section 3 for the details).

Our main results are as follows (see also Section 8 for more details):

- First demonstration of emitter fluorescence antibunching in liquid crystal hosts (Fig. 1);
- We demonstrated, for the first time to our knowledge, *a definite* polarization in fluorescence from *single* emitters (dye molecules) at room temperature (Figs 2-3). A planar-aligned, nematic liquid-crystal host provides definite alignment of single dye molecules in a preferred direction. The liquid crystal oligomer with low fluorescence background has been synthesized at the University of Rochester. We also used doped monomeric nematic liquid crystal with the same result.

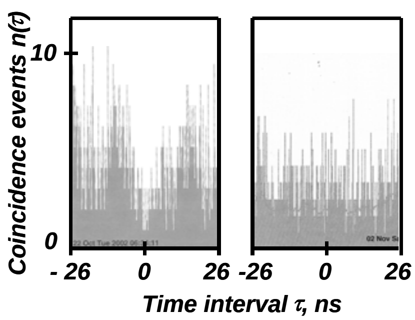

Fig. 1. Left histogram exhibits a dip at τ = 0 indicating photon antibunching in the fluorescence of the single molecule; no antibunching is observed in the fluorescence from the assembly of several molecules (right histogram).

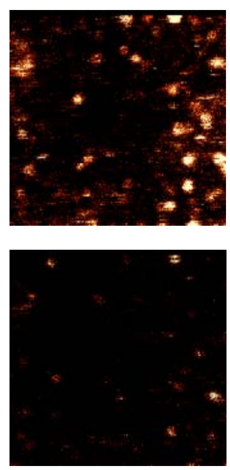

Fig. 2. Confocal fluorescence microscopy images of DiIC$_{18}$(3) dye single-molecule fluorescence in planar aligned glassy nematic liquid crystal host (10 μm x 10 μm scan): Top – polarization perpendicular to the alignment direction; bottom – parallel polarization. (*See details in Section 8*).

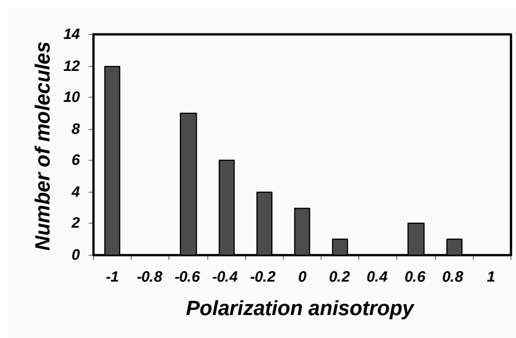

Fig. 3. The histogram of polarization anisotropy of 38 molecules of DiIC$_{18}$(3) dye in planar aligned glassy nematic liquid crystal host (*See details in Section 8*).

- Cw excitation of the embedded molecules *without bleaching* for periods of more than one hour (Fig. 4). This was achieved by special preparation of the liquid crystals;
- Preparation of 1-D photonic crystals for visible and near-infrared in chiral nematic liquid crystals (Fig. 5, left and center);
- Preparation of 2-D photonic crystals in defects of chiral nematic liquid crystals (Fig. 5, right).

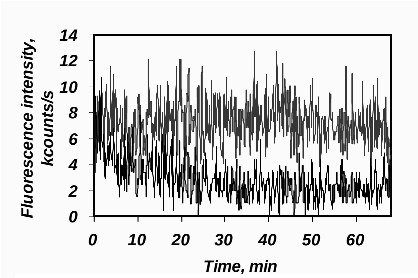

Fig. 4. No dye bleaching was observed in the oxygen-depleted liquid crystal (cyanobiphenyl 5CB) over the course of more than one hour (upper curve) in difference with doped 5CB as received (lower curve).

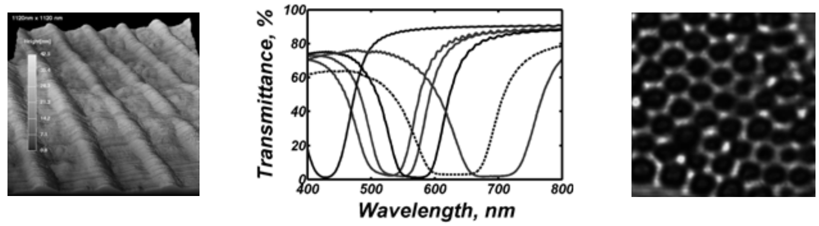

Fig. 5. Left: Perspective view of the AFM-topographical image of a planar-aligned chiral liquid crystal oligomer (1.12μm x 1.12μm scan); Center: Selective reflection curves of left-handed circularly polarized light from 1-D photonic band gap chiral liquid crystal oligomers; Right: Near-field optical image of 2-D self-assembly of a planar-aligned chiral liquid crystal oligomer (5 μm x 5 μm scan)

- Building a new confocal microscope (Fig. 6) [in addition to one with cw-laser excitation (Fig. 7)] for single-molecule imaging and photon statistics measurement with *pulsed* laser excitation (single-photon source on demand) using an NSF instrumentation grant. Single-molecule fluorescence imaging was carried out using a new microscope, electronics and software (Figs 8 and 9). This work was carried out in collaboration with Prof. L. Novotny, University of Rochester and Dr. A. Lieb, University of Basel, Switzerland.

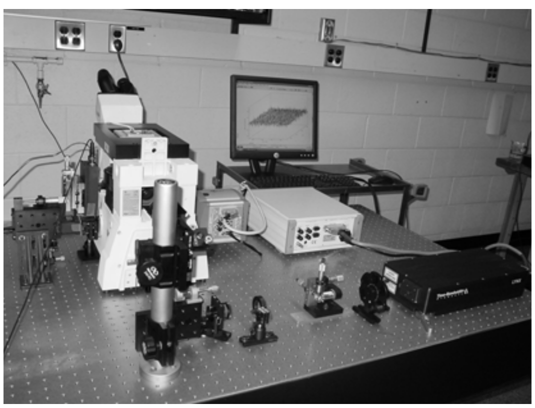

Fig.6. Photograph of the new single-photon-on-demand generation and characterization unit with pulsed laser excitation.

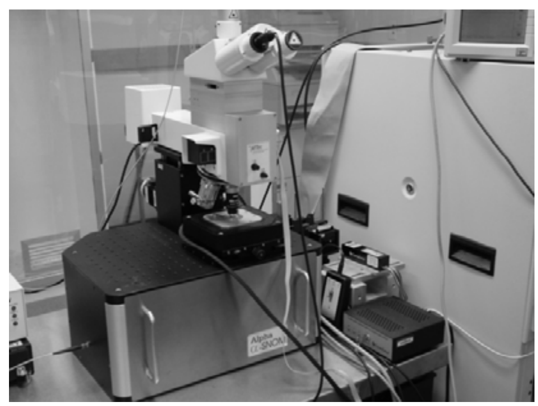

Fig.7. Photograph of a single-photon source setup with cw laser excitation, based on α -SNOM unit (WiTech Inc.) operating in a confocal transmission mode.

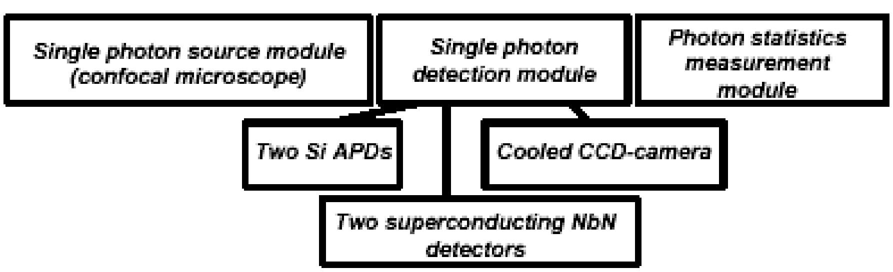

Fig. 8. The main modules of a new single-photon on demand generation and characterization unit. One port of a new confocal microscope is left for 1. 55 μm detectors. For this purpose we are planning to use superconducting NbN detectors of Prof. Sobolewski to record fluorescence from single PbSe colloidal quantum dots.

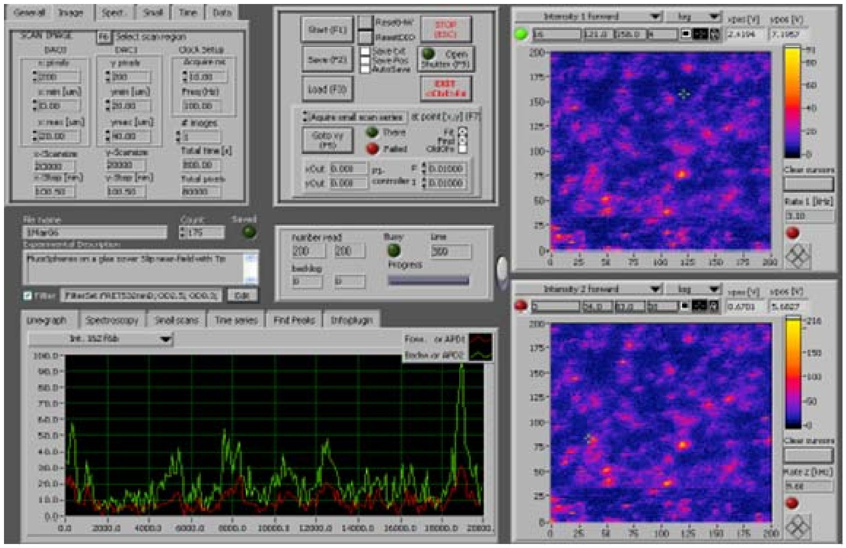

Fig. 9. Single DiI dye-molecule fluorescence imaging on a new single-photon generation and characterization unit with pulsed, 75 MHz repetition rate excitation at 532-nm (20 µm x 20 µm scan).

- 1-D photonic band-gap structures in low-fluorescence-background *monomeric* cholesteric hosts doped with single fluorescence emitters (dye molecules and quantum dots) were prepared with bandgap matching the emitter fluorescence band We succeeded in single-dye-molecule fluorescence imaging of these structures, overcoming a challenge with host fluorescence (Fig. 10). Our current single-photon source efficiency in such structures is ~10%. We are working on its further improvement;

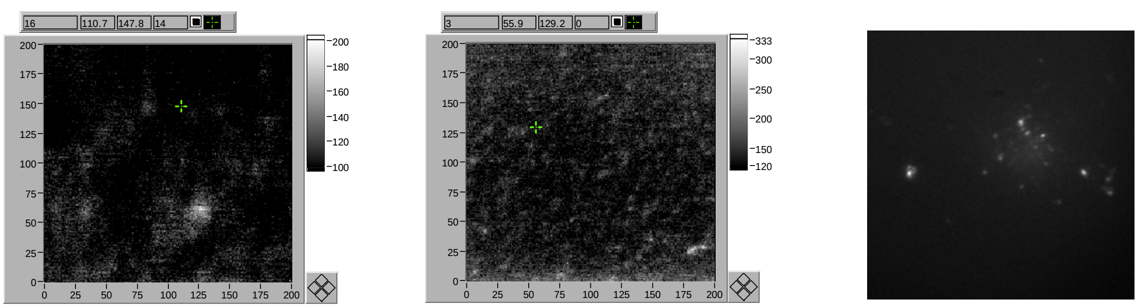

Figure 10. Single-molecule fluorescence imaging of DiI dye in liquid crystal hosts: left and center images (1-D photonic bandgap cholesteric structure) – raster scan of 10 µm x 10 µm (left) and 40 µm x 40 µm (center) and electron multiplying (EM) CCD-camera image of dye-doped planar-aligned nematic layer (right).

- We proved experimentally that single CdSe colloidal semiconductor quantum dots can fluoresce in both monomeric and oligomeric liquid crystal hosts (Fig. 11). This is a major step toward a deterministically polarized and efficient single-photon source for telecommunication wavelength using PbSe quantum dots emitting at 1.55 µm;
- A course was prepared and is being taught on Quantum Optics and Quantum Information Laboratory using modules of the new instrument in two laboratory experiments.

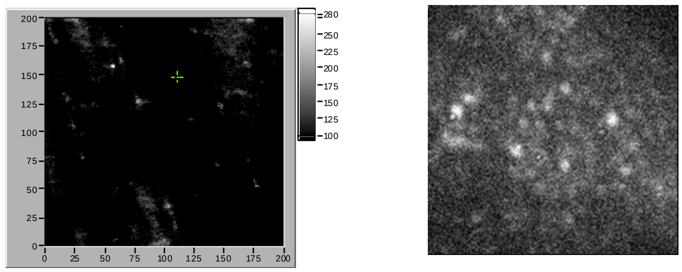

Figure 11. Single-quantum dot fluorescence imaging in 1-D photonoc bandgap cholesteric liquid crystal host by 40 μm x 40 μm raster scan (left) and EM-CCD-camera (right).

3. Publications

3.1. Papers published (Number -18)

Peer-reviewed journals (Number -16):

1. “Room temperature source of single photons of definite polarization”, S. G. Lukishova, A. W. Schmid, R. Knox, P. Freivald, L. Bissell, R.W. Boyd, C.R. Stroud, Jr, K. L. Marshall, *J. Modern Optics, Special issue on Single Photon: Detectors, Applications and Measurements Methods*, 13 pages, will be published in 2006.
2. “Near-field optical microscopy of defects in cholesteric oligomeric liquid crystal films”, S.G. Lukishova. and A.W. Schmid, *Molec. Cryst. Liq. Cryst.*, **454**, 417-423 (2006).
3. “Single-photon source for quantum information based on single dye molecule fluorescence in liquid crystal host”, S.G. Lukishova, A.W. Schmid, R.P. Knox, P. Freivald, A. McNamara, R.W. Boyd, C.R. Stroud, Jr., K.L. Marshall, *Mol. Cryst. Liq. Cryst.*, **454**, 403-416 (2006).
4. **“**Far-field patterns from dye-doped planar-aligned nematic liquid crystals under nanosecond laser irradiation”, S.G. Lukishova, N. Lepeshkin, R.W. Boyd, K. Marshall, *Molec. Cryst. Liq. Cryst.*, **453**, 393-401 (2006).
5. **“**Phase-difference equations: a calculus for quantum revivals”, D. L. Aronstein and C. R. Stroud, Jr., *Laser Physics*, **15**, 1496 (2005).
6. “Deterministic secure communications using two-mode squeezed states”, A.M. Marino and C.R. Stroud, Jr., *Phys. Rev. A* **74**, 022315 (2006).
7. “Robust multipartite multilevel quantum protocols”, H. Nihira and C. R. Stroud, Jr. , *Phys. Rev. A* **72**, 022337 (2005).
8. “Transfer of amplitude and phase modulation to a different wavelength using coherently prepared sodium vapor”, R. S. Bennink, A. M. Marino, V. Wong, R. W. Boyd, and C. R. Stroud, Jr. , *Phys. Rev. A* **72**, 023827 (2005).
9. “Dye-doped cholesteric-liquid-crystal room-temperature single photon source”, S.G. Lukishova, A.W. Schmid, Ch. M. Supranowitz, N. Lippa, A. J. McNamara, R.W. Boyd, and C.R. Stroud, *J. of Modern Optics, Special Issue on Single Photon: Detectors, Applications and Measurements Methods,* **51**, No 9-10, 1535-1547 (2004).
10. “Pas de Deux for Atomic Electrons”, C. R. Stroud, Jr., *Science* **303**, 778 (2004).

11. "Pixel Entanglement: Experimental Realization of Optically Entangled d=3 and d=6 Qudits", M.N. O'Sullivan-Hale, I. Ali Khan, R.W. Boyd, J.C. Howell, *Phys. Rev. Lett*. **94**, 220501 (2005).
12. "Realization of the Einstein-Podolsky-Rosen Paradox Using Momentum-and Position-Entangled Photons from Spontaneous Parametric Down Conversion", J.C. Howell, R.S. Bennink, S.J. Bentley, and R.W. Boyd, *Phys. Rev. Lett.* **92**, 210403 (2004).
13. "Quantum and Classical Coincidence Imaging", R.S. Bennink, S.J. Bentley, R.W. Boyd, and J.C. Howell, *Phys. Rev. Lett.*, **92**, 033601 (2004).
14. "Image formation using quantum-entangled photons", R.W. Boyd, R.S. Bennink, S.J. Bentley, J.C. Howell, published in the "Optics in 2004" section of *Optics and Photonics News*, December, 2004.
15. "Room-temperature single photon source: single dye molecule fluorescence in liquid crystal host", S.G. Lukishova, A.W. Schmid, A.J. McNamara, R.W. Boyd, and C.R. Stroud, *IEEE J. of Selected Topics in Quantum Electronics, Special issue on Quantum Internet Technologies,* **9**, N 6, 1512-1518 (2003).
16. "Cumulative birefringence effects of nanosecond laser pulses in dye-doped planar nematic liquid crystal layers", S.G. Lukishova, R.W. Boyd, N. Lepeshkin, and K.L. Marshall, *J. Nonl. Opt. Phys. & Mater., Special issue on Novel Optical Materials and Applications*, **11**, December, 341-350 (2002).

Non-peer-reviewed journals (Number –2):

1. "Deterministically polarized fluorescence from uniaxially aligned single dye molecules", S. G. Lukishova, A. W. Schmid, R. Knox, P. Freivald, L. Bissell, R.W. Boyd, C.R. Stroud, Jr, K. L. Marshall, *LLE Review*, *Quarterly Report*, *University of Rochester, Laboratory for Laser Energetics,* January-March , DOE/SF/19460-667, **106**, 102-107 (2006).
2. "Demonstration of a room-temperature single-photon source for quantum information: single-dye-molecule fluorescence in a cholesteric liquid crystal host", S.G. Lukishova, A.W. Schmid, A. J. McNamara, R.W. Boyd, and C.R. Stroud, *LLE Review*, *Quarterly Report*, DOE/SF/19460-485, Laboratory for Laser Energetics, University of Rochester, **94**, Jan-March, 97-106 (2003).

3.2. Conference and meeting presentations (Number –30):

Peer reviewed conference publications (Number -18):

1. "Deterministically polarized, room temperature source of single photons based on single-emitter fluorescence in aligned liquid crystal hosts", S.G. Lukishova, A.W. Schmid, R.P. Knox, P. Freivald, L. Bissell. R.W. Boyd, C.R. Stroud, Jr, K.L. Marshall, *Technical Digest, CLEO/QELS Conference*, May 21-26, 2006, Long Beach, CA, paper JWB97, 2 pages, 2006.
2. "Deterministically polarized, room temperature source of single photons", S.G. Lukishova, A.W. Schmid, R. Knox, P. Freivald, S. Schrauth, L. Bissell, R.W. Boyd, C.R. Stroud, Jr, K.L. Marshall, *Second Single-Photon Workshop (SPW2005*): *Sources, Detectors, Applications and Measurement Methods*, October 24-26, 2005, Teddington, UK, pp. 15-16 (2005).
3. "Quantum optics and quantum information teaching laboratory at the Institute of Optics, University of Rochester", S.G. Lukishova, C.R. Stroud, Jr., A.K. Jha, L. Elgin, S. Schrauth, L. Bissell, *Second Single-Photon Workshop (SPW2005): Sources, Detectors, Applications and Measurement Methods*, October 24-26, 2005, Teddington, UK, pp. 83-84 (2005).

4. "Oligofluorene as a new high-performance dye for cholesteric liquid crystal lasers", K. Dolgaleva, S. K. Wei, A. Trajkovska, S. Lukishova, R.W. Boyd, S.- H. Chen, *Technical Digest*, *Frontiers in Optics/Laser Science* 2006, October 8-12, 2006, Rochester, NY, paper FThD4.

5. "Cholesteric liquid crystal laser using an oligofluorene for high performance and spectral purity", K.-H. Wei, K. Dolgaleva, A. Trajkovska, S. Lukishova, R.W. Boyd, S.H. Chen, *Organic Photonics and Electronics*, October 9-11, 2006, Rochester, NY, paper OPTuD16.

6. Single-photon source for quantum information based on single dye molecule fluorescence in liquid crystal host, S.G. Lukishova, A.W. Schmid, R.P. Knox, P. Freivald, C.M. Supranowitz, A.J. McNamara, R.W. Boyd, C.R. Stroud, Jr., K.L. Marshall, *Book of Abstracts of the 11th International Topical Meeting on Optics of Liquid Crystals* (2-7 October 2005, Sand Key, Florida), p. 26 (2005).

7. Feedback-free hexagon pattern formation with liquid crystals and isotropic liquids, S.G. Lukishova, N. Lepeshkin, R.W. Boyd, K.L. Marshall, *Book of Abstracts of the 11th International Topical Meeting on Optics of Liquid Crystals* (2-7 October 2005, Sand Key, Florida), pp. 42-43 (2005).

8. Near-field optical microscopy of cholesteric oligomeric liquid crystal layers, S.G. Lukishova and A.W. Schmid, *Book of Abstracts of the 11th International Topical Meeting on Optics of Liquid Crystals* (2-7 October 2005, Sand Key, Florida), pp. 112-113 (2005).

9. Deterministically polarized fluorescence from single dye molecules aligned in liquid crystal host, S.G. Lukishova, A.W. Schmid, R.P. Knox, P. Freivald, R.W. Boyd, C.R. Stroud, Jr, K.L. Marshall, *Technical Digest*, paperJWH1-2, *IQEC 2005* (11-15 July 2005, Tokyo, Japan).

10. Deterministically polarized fluorescence from single dye molecules aligned in liquid crystal host, S.G. Lukishova, A.W. Schmid, R.P. Knox, P. Freivald, R.W. Boyd, C.R. Stroud, Jr, K.L. Marshall, *Technical Digest*, paper QTuE6, *QELS 2005,* May 2005. Baltimore, MD).

11. Single-beam light-induced phenomena in dye-doped liquids and liquid crystals, S. Lukishova, *Technical Digest*, QWAB3-P54, *IQEC 2005* (11-15 July 2005, Tokyo, Japan).

12. "Quantum optics and quantum information teaching laboratory at the Institute of Optics, University of Rochester", S.G. Lukishova, C.R. Stroud. Jr., A.K. Jha, L. Elgin, S. Schrauth, *Technical Digest*, *Frontiers in Optics/Laser Science 2005, Forum on Education,* October 16-20, 2005, Tucson, Arizona, paper FThL2 (2 pages).

13. Deterministically polarized single-photon source, S.G. Lukishova, A.W. Schmid, R. Knox, P. Freivald, R.W. Boyd, C.R. Stroud, Jr, *Abstract Digest*, *Quantum Optics II*, *Cozumel*, Mexico, December 6-9, 2004. For presentation see website http://speckle.inaoep.mx/QOII/ppts/Lukishova.pdf.

14. Deterministically polarized, room-temperature single-photon source: single dye molecule fluorescence in liquid crystal host, S. G. Lukishova, A. W. Schmid, C. M. Supranowitz, N. Lippa, A. J. McNamara, R. W. Boyd, C. R. Stroud, *IQEC04*, San Francisco, CA (May 16-21, 2004), *Technical Digest* CD-ROM, paper IThG5, 2004.

15. *(Invited)*: Quantization via fractional revivals, C. R. Stroud, Jr., *Abstract Digest, Quantum Optics II, Cozumel*, Mexico, December 6-9, 2004.

16. Room temperature single-photon source: laser control of single dye molecule fluorescence in photonic-band-gap liquid crystal host, S.G. Lukishova, A.W. Schmid, A. J. McNamara, R.W. Boyd, and C.R. Stroud, *Quantum Electronics and Laser Science Conference QELS 2003* (Baltimore, June 2003), Technical Digest, paper JWC7.

17. Efficient room temperature single-photon source for quantum information: single dye molecule fluorescence in photonic-band-gap cholesteric liquid crystal host, S.G. Lukishova, A.W. Schmid, A. J. McNamara, R.W. Boyd, and C.R. Stroud, *Frontiers in Optics, the 87th OSA Annual meeting, Technical Digest,* paper WF6, Oct. 5-9, 2003, Tucson, Arizona.
18. "Dye-doped cholesteric liquid crystal single photon source", S.G. Lukishova, A.W. Schmid, A.J. McNamara, R.W. Boyd, and C.R. Stroud, *NIST Workshop on Single Photons: Detectors, Applications and Measurements Methods* (Gaithersburg, MD, March 31 – April 1 2003).

Non-peer-reviewed conference and meeting publications (Number – 1):

1. (*Invited*): "Feedback-free, single-beam pattern formation in dye-doped liquid crystals", N. Lepeshkin, S.G. Lukishova, R.W. Boyd, K.L. Marshall, *SPIE Optics & Photonics*, August 2006, San Diego, CA.

Papers presented at conferences and meetings but not published (Number – 11):

1. *(Invited)*: Dye-doped liquid-crystal room-temperature single photon source, S.G. Lukishova, A.W. Schmid, Ch. M. Supranowitz, A.J. McNamara, R.W. Boyd, C.r. Stroud, Jr., *Laser Science XX/Frontiers in Optics 2004*, paper LMF2, October 10-14, Rochester, NY.
2. *(Invited)*: "Quantum Optics and Quantum Information Teaching Laboratory Course**"**, S.G. Lukishova, C.R. Stroud, Jr., L. Bissell, A. K. Jha, L. Elgin, *Frontiers in Optics, Special Symposium* "Quantum Optics and *Quantum Information Teaching Experiments*", Rochester, NY, October 12 (2006).
3. "Deterministically Polarized, Room Temperature Source of Single Photons", S.G. Lukishova, *Workshop on Linear Optical Quantum Information Processing*, Baton Rouge, Louisiana, 10-12 April 2006.
4. "Quantum Optics and Quantum Information Teaching Experiments", S.G. Lukishova, *Workshop on Linear Optical Quantum Information Processing,* Baton Rouge, Louisiana, 10-12 April 2006.
5. *(Invited):* "Quantum optical engineering, C. R. Stroud, Jr., *Institute of Optics 75th Anniversary Symposium*, Optical Society of America, Rochester, NY, October, 2004.
6. "An efficient, room temperature single-photon source", S. G. Lukishova, A. W. Schmid, R. Knox, P. Freivald, L. Bissell, R.W. Boyd, C.R. Stroud, Jr, K. L. Marshall, *Industrial Associates Meeting,* April 2006, Rochester NY.
7. "Room-temperature single-photon source for quantum information based on single-dye molecules in liquid crystal hosts", S.G. Lukishova, *MRS Rochester Chapter Meeting*, April 2006, Rochester NY.
8. *(Invited)*: "Quantum weirdness", C.R. Stroud, Jr., Lecture, *Marshall University,* October 13, 2005.
9. "Room-temperature single-photon source: dye fluorescence in a liquid crystal host", S.G. Lukishova, A.W. Schmid, C.M. Supranowitz, A.J. McNamara, R.W. Boyd, C.R. Stroud, *MURI Review Meeting*, July 2003, Rochester NY, Poster.
10. "Quantum information teaching laboratory", A.K. Jha, L. Elgin, S.G. Lukishova, C.R. Stroud, *MURI Review Meeting*, July 2003, Rochester NY, poster.
11. "Quantum optics/quantum information teaching laboratory", A.K. Jha, L. Elgin, S.G. Lukishova, C.R. Stroud, *Industrial Associates Meeting*, November 2003, Rochester NY, poster.

3.3. Manuscripts submitted, but not published (Number – 2):

1. "Feedback-free, single-beam pattern formation in dye-doped liquid crystals", N. Lepeshkin, S.G. Lukishova, R.W. Boyd, K.L. Marshall, *Proceed. SPIE, Optics & Photonics*, August 2006, San Diego, CA, accepted.

2. "Vibrant Imaging: 15-fold dynamic-range enhancement by odd-halves AFM-cantilever overtones", A.W. Schmid, S.G. Lukishova, (in submission).

3.4. Book chapter (Number -1)

"Coherent Transients," J. H. Eberly and C. R. Stroud, Jr., in *Handbook of Atomic, Molecular and Atomic Physics*, editor, Gordon Drake (Springer, New York, 2005).

4. Invention (by title only)

Efficient room-temperature source of polarized single photons, S.G. Lukishova, R.W. Boyd, C.R. Stroud, *Patent Application*, filed in the US PTO on January 9, 2004 (Appl. # US 20060187993, published August 24, 2006; PCT International Patent Application No PCT/US04/00362).

5. Honors and awards

- Prof. Carlos R. Stroud received the University of Rochester School of Engineering and Applied Sciences Lifetime Achievement Award granted in recognition of distinguished achievement in research, education, and leadership;
- Two-summer-month high-school intern Nadine Lippa reached semifinalist status at the Intel Science Talent Search Program for her single-photon source project;
- Undergraduate student Christopher Supranowitz was awarded national competition Goldwater scholarship for the research on a SPS;
- Undergraduate student Christopher Supranowitz was awarded international competition SPIE scholarship for his research on a SPS;
- Undergraduate student Andrew McNamara was awarded international competition SPIE scholarship for his research on a SPS;
- National Science Foundation Material Research Instrumentation Grant (PI – C.R. Stroud, co-PIs S.G. Lukishova, W. Knox, R. Sobolewski, L. Novotny), awarded on September 15, 2004.
- Dr. Lukishova was awarded Rochester University Kauffman Foundation Initiative Grant for her course "Quantum Optics and Quantum Information Laboratory";
- Graduate student Luke Bissell was recently awarded by four-year SMART Fellowship of the US AirForce for his Ph.D work in the field of quantum information.

6. Scientific personnel

- Svetlana G. Lukishova (senior scientist);
- Robert W. Boyd (professor);
- Carlos R. Stroud (professor);
- Andrew McNamara (undergraduate student);
- Christopher Supranowitz (undergraduate student);

- Nadine Lippa (two-summer-month high school intern);
- Patrick Frievald (two-summer-month research associate);
- Russell Knox (undergraduate student);
- Luke Bissell (graduate student);
- Samuel Schrauth (undergraduate student);
- Sean White (undergraduate student);

7. Technology transfer

7.1. Quantum optics and quantum information teaching laboratory course

To attract young scientists to the field of quantum information and to give them the tools and understanding to pursue it, a new course was created, the Quantum Optics and Quantum Information Teaching Laboratory at the Institute of Optics, University of Rochester (Dr. Lukishova). Current experimental setups include for teaching laboratory experiments with written manuals: (1) Entanglement and Bell’s inequalities (Fig. 12, left); (2) Single-photon interference in a Mach-Zehnder interferometer (Fig. 12, center) and with Young’s double slit (Fig. 12. Right); (3) Confocal fluorescence microscopy of single emitters; (4) Hanbury Brown and Twiss setup and antibunching measurements. Eight students from two University departments are currently taking this course in the Fall 2006 semester.

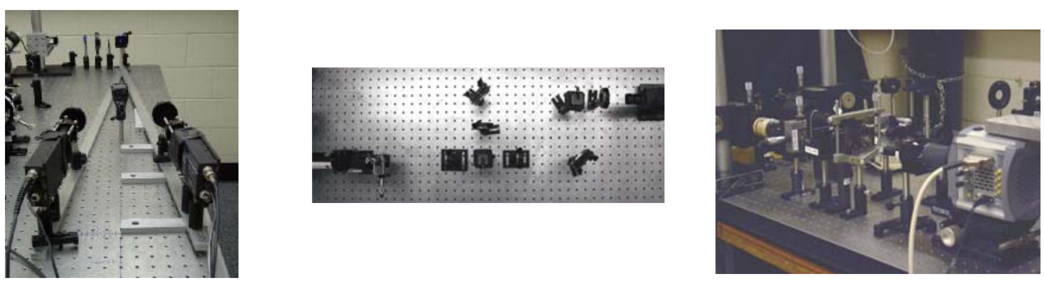

Fig. 12. Some of setups of quantum optics/quantum information teaching laboratory.

7.2. Collaboration and contacts with other researchers

- Interaction with Prof. Roman Sobolewski’s group (University of Rochester, Department of Electrical and Computer Engineering) developing fast high-repetition rate detectors for quantum cryptography. In the future, we are planning to combine our single photon source on demand with the detectors of Prof. Roman Sobolewski.
- Using CdSe quantum dots from Prof. T. Krauss (University of Rochester, Department of Chemistry). We are planning to use colloidal semiconductor PbSe quantum dots and rods prepared by his group in liquid crystal hosts for single photon source at the communication wavelegths (1.3 and 1.55 microns).
- Contacts with Dr. Chip Elliott, BBN-Technologies, Cambridge, MA building the world's first Quantum Key Distribution network with the information about our single photon source. Dr. Elliott submitted a collaborative letter to the current NSF Material Research Instrumentation project.
- Collaboration with Prof. S.H. Chen (University of Rochester, Department of Chemical Engineering) whose group synthesized nematic liquid crystal oligomers for polarized single photon source.
- Contacts with Dr. D. Subacius, MagiQ, Somerville, MA, building commercial quantum cryptography system with the information about our SPS.

8. Detailed account of the accomplishments under current ARO project

Year 1 Accomplishments

This project aims at developing a highly efficient source of deterministically polarized single photons on demand for quantum information using single-dye-molecule fluorescence in liquid crystal hosts. During this first funding period our work on this project has been devoted to achieving three goals: (1) demonstration and characterization of a single photon source; (2) preparation and characterization of 1-D-photonic band-gap structures in dye-doped cholesteric oligomers and monomers with band-gaps in various spectral regions from 400 nm to 2200 nm; (3) avoiding dye bleaching.

- A robust room-temperature single-photon source based on fluorescence from a single dye molecule was demonstrated (see antibunching histogram in Fig. 1) for the first time for liquid crystal hosts and characterized. In the case of single terrylene molecule fluorescence in a purified Wacker oligomer liquid crystal host, the probability for single photon emission into an optical fiber core is ~ 4%. The rate of two-photon pulses is four times lower than for Poissonian light.
- For further increasing single photon source (SPS) efficiency, several tens of doped planar-aligned cholesteric liquid crystal samples with band-gaps in different spectral regions from 430 nm to 2200 nm were prepared. Fig. 5, center shows transmittance of various Wacker oligomer cholesteric-liquid-crystal layers with 1-D photonic band-gaps versus wavelength in *left-handed* circularly polarized light. Similar results were achieved with the E7 + CB15 monomeric mixtures in *right-handed* circularly polarized light. To prepare the 1-D photonic band-gap structures, three different planar alignment procedures for liquid crystals were used: (i) substrate shearing, (ii) buffing, and (iii) photoalignment. The samples were prepared with a high level of homogeneity, low scattering, and high ratio between maximum and minimum transmittance. All these parameters are important for SPS operation.
- Using saturation of monomeric liquid crystals with helium or argon, terrylene dye bleaching was avoided for more than 1 hour of cw, 532 nm irradiation (Fig. 4).
- 2-D hexagonal structures (Fig. 5, right) have been prepared and characterized using cholesteric liquid crystal oligomer solutions.
- During the characterization of 2-D hexagonal structures in cholesteric oligomers, we developed a new method of AFM with a contrast enhancement using high harmonics and odd-halves AFM-cantilever overtones both in tapping and lateral mode. It is well known that in imaging contrast is very often more important than resolution. In addition, up to 15 times dynamic range enhancement over topography at 13/2 harmonic overtone was stimulated by a second cantilever eigenmode. This work was made in collaboration with Dr. A. Schmid, Laboratory for Laser Energetics, University of Rochester.

Year 2 Accomplishments

During this reporting period our work on this project has been devoted to obtain a source of *deterministically* polarized single photons using single-molecule fluorescence in planar aligned *nematic* liquid crystal hosts (1). Two other tasks have been accomplished: (2) Diminishing bleaching of dye molecules in glassy nematic liquid crystals; (3) Theoretical modeling of single-molecule fluorescence in photonic bandgap cholesteric liquid crystal hosts.

- We demonstrated, for the first time to our knowledge, *deterministically* linearly polarized fluorescence from single dye molecules. This work was made in a collaboration with Prof. S.H. Chen, Department of Chemical

Engineering, University of Rochester whose group synthesized liquid crystal oligomer with *low fluorescence* background. A nematic liquid crystal state of this material can be preserved at room temperature by slowly cooling it to the glassy state with frozen molecular orientation. We prepared the oriented films of this dye-doped glassy nematic liquid crystal aligned by a polarized UV-light (photoalignment) with one direction (~100-nm thickness).

Figs 2 and 3 show deterministically polarized fluorescence from single molecules of $DiIC_{18}(3)$ dye in planar aligned glassy nematic liquid crystal host, using a polarizing beamsplitter for separation of two polarization states. The polarization of the fluorescence of single molecules of this dye is predominantly in the direction perpendicular to the alignment of liquid crystal molecules. (Polarization anisotropy is defined here $P = (I_{par} - I_{perp})/I_{par} + I_{perp})$, where I_{par} and I_{perp} are fluorescence intensities for polarization parallel and perpendicular to alignment direction). The same sign of polarization anisotropy we obtained with high (~0.5% by weight) concentration of the same dye in aligned glassy nematic liquid crystal. Spectrofluorimeter measurements of fluorescence from this sample for two different polarizations depicted in Fig. 13 (P ~ -0.6). The absorbing/emitting dipole direction of this dye is nearly perpendicular to the length of the molecule which aligns parallel to the liquid crystal molecules. Fig. 14 shows a molecular structure of used $DiIC_{18}(3)$ dye.

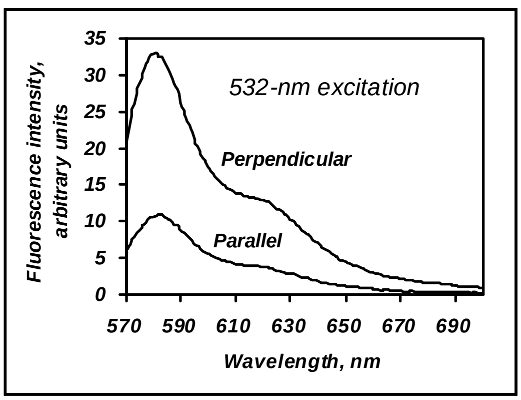

Fig. 13. Fluorescence spectra in planar aligned glassy nematic liquid crystal host for different polarizations under the excitation with a nonpolarized, 532 nm light.

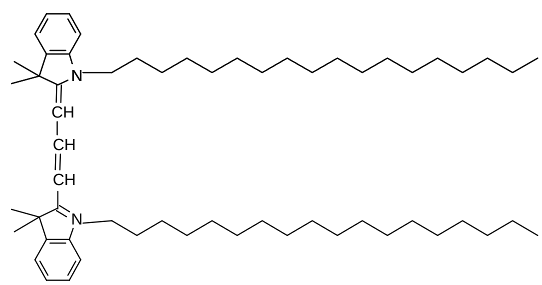

Fig. 14. Molecular structure of $DiIC_{18}(3)$ dye used in our experiments.

Year 3 Accomplishments

In third-year reporting period, we had two main goals: (1) building a single-photon source *on demand* using a NSF instrumentation grant; (2) investigating single-molecule fluorescence in 1-D photonic bandgap cholesteric liquid crystal structures for increasing SPS efficiency.

- For this purpose, we built a new robust single-photon-on-demand-generation and characterization unit using a *pulsed* laser (in addition to the cw-laser instrument), providing both single-emitter fluorescence imaging and photon-statistic measurements.
- We also made 1-D photonic band-gap structures in *low-fluorescence-background* cholesteric hosts doped with fluorescence emitters. This work was carried out in collaboration with Prof. S.H. Chen (Department of Chemical Engineering, University of Rochester) and Cornerstone Research Group (Dayton, OH) whose

groups synthesized two types of liquid crystal oligomers with low fluorescence background. Chiral compound can be mixed in solution with a nematic compound in different proportions, and after several procedures (spin-coating on a photoaligned polymer, heating and slow annealing) 1-D photonic band-gap cholesteric structures can be frozen in a glassy state. Band gap position depends on the relative concentration of the compounds. Transmission in the band center depends on the glassy-layer thickness. We succeeded in single-molecule fluorescence imaging of these structures, overcoming a challenge with host fluorescence.

- Theoretical analysis of spontaneous emission in cholesterics with a wide-angle approximation was made. Ph.D student Anand Jha worked on theoretical modeling of the spontaneous emission from a fluorescent emitter inside a cholesteric photonic band-gap layer. His modeling takes into account the high-angle modes which may influence fluorescence lifetime. This work is being carried out in collaboration with Prof. J. Dowling (Louisiana State University).

Year 4 Accomplishments (no-cost extension period)

In fourth-year reporting period, we had two main goals: (1) developing and optimizing the *single-photon-on-demand generation and characterization unit* built during the third year for the visible spectral range and its preparation for use at telecommunication wavelengths; (2) using CdSe colloidal semiconductor quantum dots as fluorescence emitters in liquid crystal hosts; (3) investigating single-emitter fluorescence in monomeric (fluid like) liquid crystals (both in planar-aligned nematic and 1-D photonic bandgap cholesteric liquid crystal structures; (4) increasing efficiency of the single-photon source; (5) using SPS-instrument for student teaching.

- Our experience with single DiI dye molecule fluorescence in liquid crystal hosts at pulsed laser excitation (76 MHz pulse repetition rate) shows much higher stability of the dye fluorescence in *liquid crystal host* (excitation periods of ~ 20 min or more without bleaching) even without the oxygen depletion procedure. At the same time single dye molecules on a bare glass surface are bleached very fast (~ 30 s or up to 3 min. with a polymethylmethacrylate coating) at comparable incident intensity;
- 1-D photonic band-gap structures in low-fluorescence-background *monomeric* cholesteric hosts doped with single fluorescence emitters (dye molecules and quantum dots) were prepared with bandgap matching the emitter fluorescence band. We succeeded in single-dye-molecule fluorescence imaging of these structures, overcoming a challenge with host fluorescence. Our current single-photon source efficiency in such structures is ~10%;
- We proved experimentally that single CdSe colloidal semiconductor quantum dots can fluoresce in both monomeric and oligomeric liquid crystal hosts. This is a major step toward a deterministically polarized and efficient single-photon source for telecommunication wavelength using PbSe quantum dots emitting at 1.55 μm;
- Our previous results on deterministically polarized fluorescence from single molecules of DiI dye in *oligomeric* nematic liquid crystal hosts were confirmed for *monomeric* liquid crystal hosts;
- A course was prepared and is being taught on Quantum Optics and Quantum Information Laboratory using modules of the new instrument in two laboratory experiments.